
Attribute and Semiquantitative Measurements

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20.1 INTRODUCTION

Nuclear material measurements are usually quantitative assays where the measurement goal is to fix a numerical value on the amount of nuclear material present. The assays are performed with the highest accuracy and precision possible, and prior knowledge about the samples may be extensive. There are however a number of measurement challenges that can be met with more qualitative information on samples about which prior knowledge may vary widely. Some examples follow:

- characterization of unlabeled or mislabeled samples
- go/no-go determination of nuclear material content for recovery, burial, transport, or criticality safety
- rapid inventory verification to check consistency of declared values
- confirmation of shipper values by the receiver
- location of nuclear material holdup
- process monitoring
- control of material movement.

Most of these tasks can be accomplished with qualitative or semiquantitative measurements that are rapid enough to save time, money, and personnel exposure.

Nondestructive assay techniques are well suited to these types of measurements because they are usually fast, nonintrusive, and capable of measuring the package as a whole. If the nondestructive measurement is careful and accurate, it may be considered a *material assay*. If the measurement is completely qualitative and only determines some signature, fingerprint, quality, or characteristic of the material, it may be considered an *attribute measurement*. Between these extremes are semiquantitative measurements such as waste characterization, monitoring of material movement, rapid inventory verification, and identification and measurement of material holdup. These semiquantitative measurements are often very important to the day-to-day operation of nuclear fuel-cycle facilities.

Section 20.2 summarizes nuclear material attributes and how they can be measured. The remainder of the chapter discusses semiquantitative measurements of waste (Section 20.3), confirmatory measurements for inventory verification and shipping (Section 20.4), and holdup measurements (Sections 20.5 and 20.6). These semiquantitative measurements are more than attribute measurements but less than full quantitative assays. For discussions of two other measurement problems that fall into the

category of semiquantitative measurements, see Chapter 18 on irradiated fuel assay and Chapter 19 on portal monitoring.

20.2 MEASUREMENT OF NUCLEAR MATERIAL ATTRIBUTES

The most fundamental task in measuring nuclear material attributes is simply to identify the presence or absence of nuclear material in a sample. In this regard the term "nuclear material" refers to all forms and combinations of uranium and plutonium, to radioactive sources, and to americium, thorium, and other radioactive elements. The primary radiation attributes (regardless of material type) are listed below:

- alpha radiation
- beta radiation
- gamma radiation
- infrared radiation (heat)
- total neutron radiation
- coincident neutron radiation
- high fission cross section for thermal neutrons (yielding prompt and delayed gamma rays and neutrons).

(Information on the radiation emission rates of these attributes is summarized in Chapters 1, 11, and 21 and in Refs. 1 and 2.)

Nuclear material in elemental form is also very dense and strongly attenuates gamma radiation. A further attribute of uranium and plutonium is the discontinuities in their x-ray absorption cross section at the K- and L_{III}-absorption edges (Chapter 9). Of all the attributes listed above, only the gamma-ray transmissions at the absorption edges provide a unique identification. In practice, however, the fissile character of uranium and plutonium is essentially unique, since fissile isotopes of other elements would not be expected in fuel-cycle facilities. Gamma-ray spectroscopy also provides an unambiguous identification, especially if the spectra are measured with high resolution. Although the other attributes mentioned are necessary features of nuclear material, they are not sufficient for unique identification.

In a full-fledged nuclear material assay, almost all of the attributes cited above are measured at one time or another. A simple way to view attributes measurements is to regard them as incomplete assays. The data are taken in the same way as for complete assays but the measurements are made more quickly, with less precision, and often without any use of the absolute calibration of the instrument. Even semiquantitative confirmatory or verification measurements may involve only a determination of the relative magnitude of the attribute responses from sample to sample. Table 20-1 summarizes the measurement instruments that are commonly available in nuclear facilities and the attributes they can reveal. Some active assay instruments are included for completeness.

Attribute measurements can be a very effective tool for characterizing, verifying, or monitoring nuclear material. Measured one at a time, nuclear material attributes provide simple answers to inventory questions. Measured in combination, they can provide very reliable or even unique information with a minimum of effort.

Table 20-1. Measurement instruments and the attributes they reveal

Instrument	Attribute
Visual inspection	packaging, history, color
Scales	weight, density
Alpha counter	presence of alpha particles, contamination
Geiger counter	gross beta/gamma activity; presence of U, Pu, or Am
Gamma spectrometer	gamma-ray spectrum; U, Pu signature; enrichment; burnup
Radiograph	density, distribution, shape
Densitometer	density; x-ray absorption edges; U, Pu signature
Calorimeter	heat output; presence of high alpha activity; warmth implies Pu, Am
Passive total neutron counter	neutron emission; presence of spontaneous fissions or (α, n) reactions
Passive neutron coincidence counter	spontaneous or induced fissions; presence of Pu or Cf likely
Active neutron coincidence counter	induced fissions; presence of U or Pu likely
Californium shuffler	delayed neutrons from induced fissions; presence of U or Pu likely
Fuel-rod scanner	delayed neutrons or gamma rays; presence of U or Pu likely

20.3 QUANTITATIVE SCREENING OF WASTE

20.3.1 Purpose

Nuclear fuel-cycle facilities often generate large quantities of waste that is only slightly contaminated or that is assumed to be contaminated because of its proximity to other materials. This type of waste is usually packaged in 55-gal. drums or larger containers and sent to retrievable storage in shallow burial sites. To minimize the volume of waste that will ultimately have to be retrieved, it is important to determine the level of radioactivity in the waste at the point of generation. Current regulations permit burial of waste in nonretrievable storage if the level of radioactivity is below 100 nCi/g of waste. (The average level of radioactivity in US soil is about 10 nCi/g. Until recently, the cutoff for permanent burial was set at this limit.) The purpose of nondestructive screening of low-level waste is to supplement or replace administrative controls for waste sorting at the 100-nCi/g level. Present experience suggests that the volume of nonretrievable waste can then be reduced by a factor of 10 or more.

Nondestructive measurement of low-level waste is difficult because the containers used are large [ranging from 2-cu-ft boxes (57 L) to 55-gal. drums (208 L) to 4- by 4- by 7-ft crates (3300 L)] and the quantities of nuclear material involved are small. A radiation level of 100 nCi/g is equivalent to about 160 mg of ^{239}Pu , 1 g of ^{233}U , or 4.4 kg of ^{235}U in 100 kg of waste. Because of the large container sizes and the low level of radioactivity,

nondestructive measurements emphasize sensitivity rather than accuracy. The measurement goal is often 1- to 10-nCi/g sensitivity and $\pm 20\%$ accuracy. This level of accuracy is considered sufficient for waste screening, with the proviso that large systematic underestimations must be avoided so that significant quantities of recoverable nuclear material are not lost and so that the actual quantity of buried nuclear material does not exceed criticality safety guidelines.

20.3.2 Gamma-Ray and Neutron Sensitivities

Table 20-2 summarizes the approximate detectability limits of nondestructive assay techniques for ^{235}U and plutonium (10% ^{240}Pu , 90% ^{239}Pu) (Refs. 3 through 5). The detectability limits are given for 1000-s measurements with the signal being three standard deviations above background (99% confidence level). The detectability limit is a function of the detector response per gram of nuclear material and of the ambient background (as given by Equations 15-7 and 15-8 in Chapter 15). The limits quoted in Table 20-2 are based on reasonable estimates for background contributions. The detectability limit is also a function of the detector size and efficiency. The limits in Table 20-2 are measured or extrapolated values for detectors that can accommodate 55-gal. drums for cases where no lead shielding is required to reduce the gamma-ray background from fission products in the waste.

In general, passive gamma-ray counting by segmented scanning is not quite sensitive enough to screen waste at the 100-nCi/g level. For contamination levels above 1 g, segmented gamma-ray scanning is the most reliable technique for quantitative measurements of ^{235}U and ^{239}Pu up to the limit of penetrability of the 186- and 414-keV gamma rays. For 57-L cardboard boxes containing waste with an average density of 0.1 g/cm^3 , passive counting of L x rays has a demonstrated sensitivity of less than 1 nCi/g (Ref. 6).

Although passive gamma-ray measurements of nuclear waste are usually biased low because of gamma-ray self-attenuation, passive neutron measurements are usually

Table 20-2. Nuclear waste detectability limits^a (Refs. 3 through 5)

Nondestructive Assay Technique	Efficiency (%)	^{235}U	Pu (10% ^{240}Pu)
Passive gamma-ray counting	1	100 mg	100 mg
Passive thermal neutron counting	15	N/A	18 mg
Passive thermal neutron coincidence counting	15	N/A	6 mg
Passive fast neutron coincidence counting	25	N/A	300 mg
Active thermal neutron coincidence counting	15	10 g	35 g
Active fast neutron coincidence counting	25	70 mg	130 mg
Photoneutron interrogation ($3 \times 10^8 \text{ n/s}$)	0.25	8 mg	6 mg
Delayed neutron counting ($2 \times 10^9 \text{ n/s source}$)	15	6 mg	14 mg
Differential die-away neutron counting	14	1 mg	1 mg

^a1000-s counting time; signal 3σ above background; no fission-product gamma-ray shielding; nuclear material in oxide form.

biased high because of additional neutrons from (α, n) reactions. Examples of (α, n)-reaction effects are given in Section 15.5.1. Active neutron techniques are not subject to the effects of passive backgrounds if the interrogation source is strong enough. However, active techniques can be strongly biased either high or low depending on the effects of matrix moderation and absorption on the incoming and outgoing neutrons. For a given waste-screening application, the choice of measurement technique should be made on the basis of cost, simplicity, sensitivity, and penetrability.

20.4 CONFIRMATORY MEASUREMENTS

20.4.1 Purpose

The transfer and storage of unirradiated nuclear materials is a frequent and large-scale activity at many NRC and DOE facilities. Many safeguards issues arise during the process of shipping, receiving, and inventory verification. Measurements can help to confirm that (a) material has not been diverted in transit, (b) the item identification is correct, (c) there is no undue radiation hazard to workers, and (d) inventory records are credible. Such confirmatory measurements may be simpler than measurements made for accountability purposes. For example, they require less time and less unpacking or repackaging of material. They also may be more versatile. However, in general, they are less accurate. Confirmatory measurements determine such attributes as weight, gamma-ray spectrum, total neutron radiation, and enrichment that—taken as a whole—are very difficult to imitate.

When nuclear material is transferred from one facility to another, present regulations require that the receiver verify the piece count, identification, and gross weight of the items in the shipment. Normally the receiver should perform accountability measurements on the items within 10 calendar days. In practice, however, this is often difficult to achieve because of (a) limitations in the availability of personnel and nondestructive assay equipment, (b) the length of time required for performing chemical analysis and transferring shipments into and out of storage vaults, and (c) the radiation exposure to personnel during packing and unpacking. Also, difficulty in measuring a relatively small number of scrap materials can delay closing the material balance on the shipment. One safeguards approach to alleviating these problems is to make confirmatory measurements at both the shipping facility and the receiving facility with similar or identical instruments (Ref. 7). Such measurements can confirm that there are no missing, incorrect, or bogus items in the shipment.

When nuclear material is stored at a facility, present regulations require periodic inventory of the entire facility and its storage vault. Confirmatory measurements made during that time on a random sample of the inventory can help identify mislabeled items and increase the credibility of the inventory process (Ref. 8).

20.4.2 Nondestructive Assay Options

Nondestructive assay techniques are well suited for confirmatory measurements because of their speed and their ability to measure an entire item. In some cases, it is also

possible to measure the shipping container itself, although with some loss of precision and accuracy. Options based on nondestructive assay of plutonium or uranium radiation attributes are summarized in this section.

For most plutonium samples, a combination of calorimetry and gamma-ray spectroscopy provides the best available accuracy: 0.5 to 2% for homogeneous materials. However, this instrumentation is usually reserved for accountability measurements because of its relatively high cost, complexity, and low throughput. Passive gamma-ray counting of the 414-keV ^{239}Pu peak in either a far-field geometry or by segmented scanning is a simpler option for materials of low density. However, most plutonium-bearing materials that are attractive for diversion are too dense for gamma-ray counting and are best measured by passive neutron counting. The technique is relatively simple, and can sometimes be applied to 30- or 55-gal. shipping drums without unpacking their contents. The neutron well counter should have uniform efficiency over the volume of the sample. Also, the electronics deadtime should be small and well known so that count ratios can be determined accurately.

Confirmatory neutron measurements of plutonium can be based on total or coincident counting, but coincident counting is a more specific attribute. Counting times are in the range of 100 to 1000 s. Typical accuracies for quick confirmatory measurements are 1 to 10% for well-characterized materials, 25% for impure scrap, and 50% for heterogeneous materials with high (α, n) rates (Ref. 8). However, the repeatability of raw measurement results is approximately 1%. It would be very difficult technically to construct a bogus item with the same weight, total neutron count rate, and coincident neutron count rate as a real item. This is also true for heterogeneous materials with high alpha decay rates where the assay accuracy is poor but the neutron attribute measurement is quite precise.

For passive neutron measurements of plutonium, the following guidelines show how the observed count rates are related to specific material attributes:

(1) The total neutron count rate is proportional to fertile content but also depends on the (α, n) reaction rate. If the fertile content can be determined from the coincident count rate, then any "excess" total count rate can be attributed to chemical compounds or impurities.

(2) The neutron coincidence rate is proportional to fertile content, but may be enhanced by induced fissions.

(3) The coincidence/totals ratio is a function of sample self-multiplication and, indirectly, fissile content. For heterogeneous plutonium scrap with very strong (α, n) reactions, the coincidence/totals ratio may provide the best possible measure of ^{239}Pu content, perhaps within 10%, if an iterative correction for ^{240}Pu content is made (Ref. 9).

(4) The difference in coincident neutron response with and without a cadmium liner in the well counter, divided by total neutron response, is a measure of fissile content (Section 17.3.3 and Ref. 10).

Confirmatory measurements of uranium are more difficult than those of plutonium. The alpha-particle emission rates are not high enough to permit heat production measurements. Enrichment measurements are possible with the 186-keV gamma ray, but they sample only the surface of the material and require a well-collimated geometry outside of the shipping drum. Far-field gamma-ray measurements can be used for low-density materials. They have also been used to confirm high-density materials to within a factor of 2 (Ref. 8).

The measurement of bulk uranium samples requires the use of active neutron systems, with the simplest being the Active Well Coincidence Counter (AWCC) (Section 17.3.1). In the thermal mode the AWCC is appropriate for samples containing from 5 to 100 g of ^{235}U . In the fast mode the instrument is limited to samples containing 50 g of ^{235}U or more, even for 1000-s counting times, because of the high accidental coincidence background of the interrogation sources. Good coupling must be maintained between the sources and the uranium, which usually requires the use of small containers. Thus, active coincidence counting of uranium is not as versatile or as easy to apply as passive coincidence counting of plutonium.

Two specific applications of active neutron counting of uranium are summarized below:

(1) Mixed uranium/plutonium samples: The passive coincidence response is proportional to ^{240}Pu but may be enhanced by induced fission in ^{235}U . Correction for self-multiplication can compensate for induced fission but will not provide a direct measure of ^{235}U content. Determination of ^{235}U or ^{239}Pu fissile content is not practical by active coincidence counting and requires more complex active neutron systems.

(2) Highly enriched uranium in UF_6 cylinders: The coincidence/totals ratio is proportional to ^{235}U content to within 2 to 10% (Section 17.3.4).

20.4.3 Recent Experience

Several examples follow of recent confirmatory measurements at Hanford, Rocky Flats, Los Alamos, and Savannah River. The examples illustrate different approaches and different levels of accuracy; they are arranged roughly in order of increasing degrees of confirmation.

Verification of a wide variety of stored nuclear material has been obtained by performing confirmatory measurements on a random sample of the inventory (Ref. 8). Passive neutron coincidence counting of plutonium and passive gamma-ray counting of uranium in a far-field geometry were the preferred techniques. Roughly 5% of the measurements were invalidated because of poor counting statistics, unsuitable material matrices or geometries, or lack of appropriate standards. Another 5% were judged as not confirmed because of results inconsistent with those obtained earlier on similar items. For the latter 5%, a superior instrument or technique was used to perform an accountability measurement. In about half of these cases the more accurate accountability measurement verified that the original item label was indeed incorrect.

Confirmation of incoming plutonium scrap metal has been accomplished by passive neutron coincidence counting of "bird cage" shipping containers (Ref. 11). Measurement of the shipping container itself rather than the individual interior items resulted in an eightfold reduction in work hours and a thirtyfold reduction in radiation exposure. Measurement accuracy was roughly 5% (1σ) for the shipping container as a whole compared to 2.5% (1σ) for the individual items. The receiver was able to verify the incoming shipment within three working days.

Confirmatory measurements of plutonium oxide have been performed by both the shipper and the receiver, each using a neutron coincidence counter of different design (Ref. 8). The counters measured the individual cans outside of their shipping drums. No attempt was made to normalize the response of one counter to the other. Instead, the

confirmation was based on the ratio of the responses. The total neutron count ratios were consistent to 0.5% (1σ), and the coincidence count ratios were consistent to 1.5% (1σ) before and after shipment. The receiver also compared his measurement of the actual plutonium mass as obtained by coincidence counting with that obtained by calorimetry for eight batches of cans. This comparison was not as accurate, having a 4.1% (1σ) scatter. The reduced accuracy of the mass determination is attributed to differences in settling, oxide density, moisture, or isotopics between batches. For example, the coincidence response of a 1-kg plutonium oxide can will change by about 1% for a 5% change in density (see Figure 16.14 in Chapter 16).

Shipper and receiver confirmatory measurements of plutonium-bearing ash, sand, slag, crucible, and oxide have been carried out by segmented gamma scanners of different design (Ref. 8). Standards were fabricated by the shipper, calibrated on the shipper's calorimeter, and sent to the receiver. The receiver's measurements of ^{239}Pu content agreed with the shipper's measurements to within 1 to 4% (1σ).

Confirmatory measurements of impure plutonium metal and oxide have been made with two identical neutron coincidence counters that measure 30-gal.-drum shipping containers. Figure 20.1 shows cutaway views of one of the counters. The counters are the first instruments designed specifically for confirmatory measurements (Ref. 12). The design features two doors, drum rollers, a drum positioner, and void spaces in the polyethylene wall to flatten the vertical efficiency profile. Normalization of response between shipper and receiver is accomplished by exchange of ^{252}Cf sources, source measurement data, and background measurement data. The confirmatory measurements consist of three 100-s total neutron counts. Initial results provided a shipper/receiver verification within 2 to 3% for oxide and within 1% for metal (Ref. 11). There is some evidence of a small bias that may be due to settling of the contents during shipment.

20.5 NUCLEAR MATERIAL HOLDUP

The term "holdup" refers to the accumulation of nuclear material inside the processing equipment of nuclear facilities. Other common terms for such material are "hidden inventory," "normal operating loss," and "in-process inventory." The choice of terminology depends in part on the application or point of view. For example, the nuclear material that remains in the facility after the runout of all bulk product may be called "in-process inventory." The material that remains after thorough brushing, wiping, acid leaching, and rinsing may be called "fixed holdup."

Because of the high economic value of nuclear material and the need to ensure radiation safety and criticality safety and to safeguard against theft or diversion, it is important to minimize holdup, to measure or model its magnitudes, and to remove it. Holdup causes and mechanisms, holdup magnitudes, and holdup modeling and measurement techniques are discussed in the remaining sections of this chapter.

20.5.1 Causes and Mechanisms

Nuclear material tends to accumulate in cracks, pores, and regions of poor circulation within process equipment. In addition, the internal surfaces of pipes, tanks, ducts,

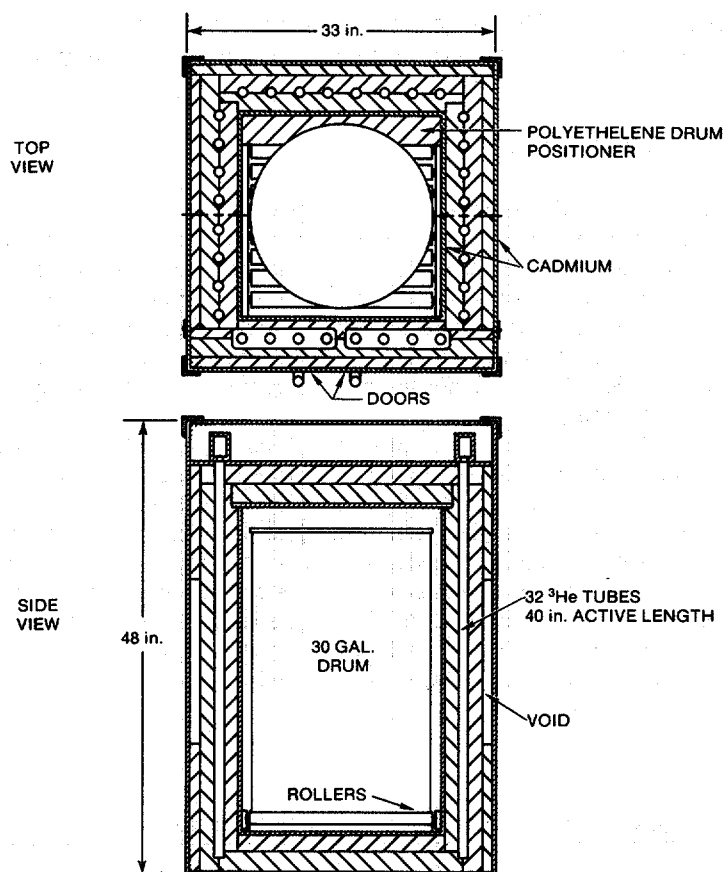


Fig. 20.1 Cutaway view of one of the Confirmatory Measurement Counters built specifically for shipper and receiver confirmatory measurements of plutonium in 30-gal. shipping drums (Refs. 11 and 12).

furnaces, gloveboxes, and other equipment can acquire appreciable deposits. When the internal surface areas are large, the total holdup can be enough to affect the plant inventory difference (Ref. 13). The amount of holdup depends on the nature of the process and on the physical form of intermediate solutions, precipitates, and powders. Also, process upsets can lead to large, rapid, and unexpected depositions of material.

Some of the mechanisms for material accumulation (Ref. 14) are summarized below:

- (1) gradual sedimentation and settling of fine particles in regions of poor circulation or low flow rate
- (2) chemical reaction of nuclear materials with interior walls or migration of the materials into the walls
- (3) solid or liquid product formation or precipitation resulting from inadvertent chemical reactions

- (4) electrostatic deposition and buildup of charged particulates
- (5) splashing, bubbling, or caking of materials resulting from unregulated chemical reactions.

20.5.2 Magnitude of Holdup

From the holdup mechanisms outlined in the preceding section it is possible to identify regions where holdup may be high. These include elbows, junctions, and seams in pipes and ducts; regions of stagnant flow or regions with turbulent flow; equipment with large interior surfaces such as Raschig-ring tanks, filters, gloveboxes, and furnaces; and wet operations with corrosive acids or high concentrations of nuclear material. The magnitude of the holdup in these regions is difficult to estimate because it depends on such factors as plant layout, frequency of process upsets, maintenance and cleanout procedures, and throughput.

Some typical holdup magnitudes observed in equipment at several uranium and plutonium processing facilities are given in Table 20-3. The numbers shown are typical of regions of high holdup only, but they suggest that extrapolation over all of the major process areas in a facility can yield tens or hundreds of kilograms of total holdup. As a fraction of total throughput, the holdup can be in the range of 0.1 to 0.2% even after thorough destructive cleaning. When nuclear material is first introduced into a new facility, the initial holdup can be 1 to 10% of the initial throughput. Because facility design can affect the amount of holdup, the Nuclear Regulatory Commission has proposed design considerations to minimize holdup (Refs. 15 through 17).

Table 20-3. Typical magnitudes of holdup in facility equipment

Gloveboxes	0 - 50 g
Gloveboxes (after destructive cleaning)	2 g/m ²
Grinders	1 - 100 g
V-blenders	1 - 50 g
Glovebox prefilters	2 - 100 g
Final filters	10 - 100 g
Equipment interiors (after routine cleaning)	10 - 50 g/m ²
Pipes (after destructive cleaning)	0.3 g/m
Ducts (no cleaning)	1 - 100 g/m
Glass columns	1 g
Annular tanks	1 - 10 g
Raschig-ring filled tanks (after rinsing)	1 - 500 g
Dissolver trays	10 - 500 g
Small calciners	5 - 50 g
Furnaces	50 - 500 g
Furnace trays	1 - 10 g
Incinerators	1000's g
Concrete spill basins	1000's g

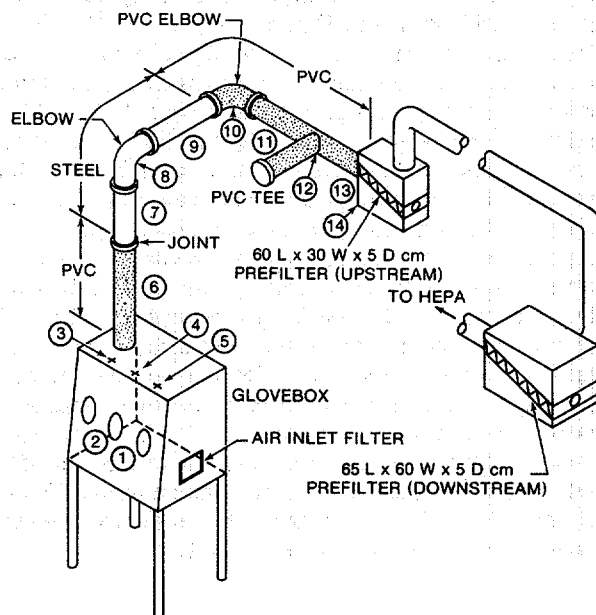
20.5.3 Statistical Modeling

Determining the location of material held up in process equipment and recovering it is very difficult. Even measuring the quantity of holdup is difficult and subject to many uncertainties (see Section 20.6). A possible alternative method for obtaining some of the holdup data required for periodic inventory is to estimate the holdup through statistical modeling (Ref. 14). This approach would begin with careful, controlled holdup measurements (either nondestructive or cleanout) of a process operation under known conditions of temperature, flow rate, throughput, and so forth. The measured holdup would be modeled as a function of the important variables. Then future holdup in this process operation could be estimated and predicted on the basis of the model.

A series of controlled holdup deposition and measurement experiments have been conducted to test the validity of this approach (Ref. 14). Figure 20.2 shows the layout of the equipment used during one such experiment designed to determine the holdup of uranium dust as a function of material characteristics, airflow rate, and dusting material. A mechanical dust-generating apparatus located inside the glovebox provided a source of airborne dust. Radioactive tracers were incorporated into the uranium oxide at a concentration of about one part per billion in order to increase the accuracy of the gamma-ray holdup measurements. Comparison with cleanout showed that the holdup measurements were accurate to about 20%.

Some of the data from the experiment are illustrated in Figure 20.3 (Ref. 14). This figure shows filter holdup increasing as a function of airflow and throughput. The holdup can be modeled as a quadratic function of throughput, as illustrated by the smooth curves in the figure. These data provide a good example of holdup that increases

Fig. 20.2 Layout of equipment used during a controlled holdup deposition and measurement study, showing the glovebox where dust is generated, ducts, filters, and the location of 14 measurement points (Ref. 14).



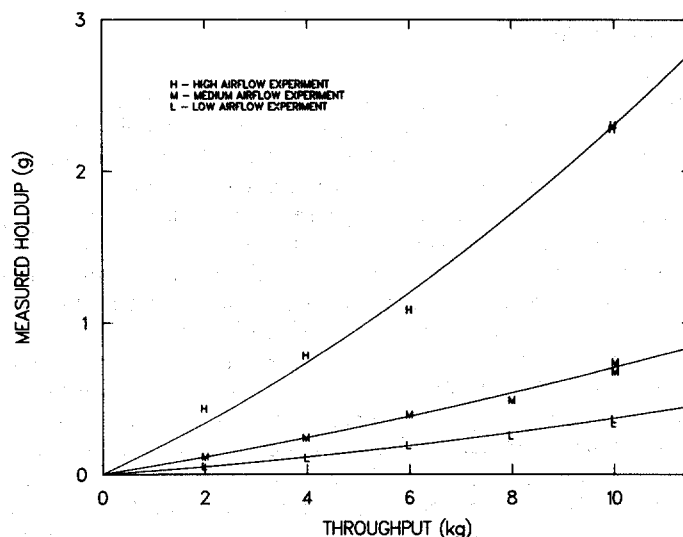


Fig. 20.3 Holdup of fine U_3O_8 powder as a function of airflow rate and total throughput as measured at the exhaust air filter (location 14 in Figure 20.2) during a controlled dust-generation experiment (Ref. 14).

steadily with increasing throughput. Another typical pattern observed in controlled experiments is illustrated in Figure 20.4 (Ref. 14). Holdup inside a calciner increased rapidly as a linear function of throughput until a coating was built up. Then the holdup fluctuated about a steady-state value until operating conditions changed. In this case, an increase in calciner operating temperature from 700°C to 900°C caused another increase in holdup. During the period of steady-state operation the data were fitted to a Kalman filter model, as indicated by the smoothed curve in the middle of the figure.

The behavior illustrated in Figure 20.4 is somewhat representative of a process facility as a whole. When nuclear material is first introduced into a new facility, holdup may build up rapidly as equipment becomes coated and cracks become filled. During subsequent years of steady operation and routine cleanout, holdup increases more slowly and may tend to approach some asymptotic value.

The controlled holdup deposition and measurement experiments described above suggest that holdup estimation models can be useful if they are based on good initial measurements, if process operation is stable, and if the data base is updated periodically. Under these conditions the frequency and number of measurements can be reduced. Controlled-holdup experiments also provide an example of the best accuracy obtainable in holdup measurements made under ideal conditions. The accuracies reported for these experiments (Ref. 14) are included in the summary of published holdup measurement accuracies given in the table in Section 20.6.7.

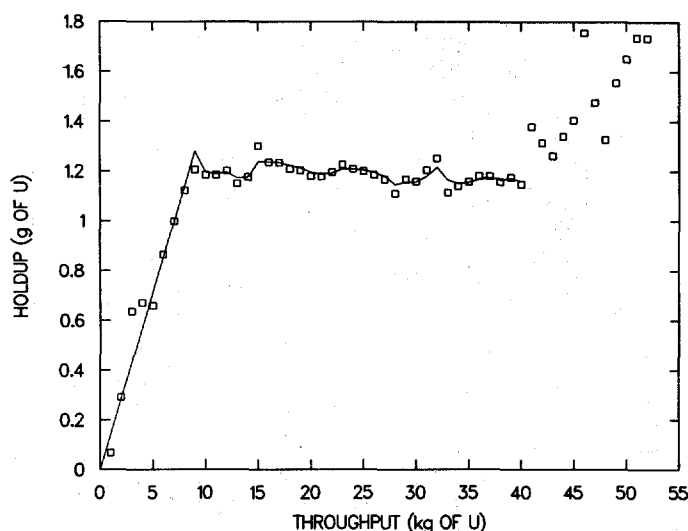


Fig. 20.4 Holdup of uranium oxide in a calciner as a function of throughput, as determined during a controlled holdup experiment. This figure is an example of the leveling out of holdup at a steady-state value after an initial buildup. At a throughput of 40 kg, an increase in furnace temperature resulted in another buildup (Ref. 14).

20.6 THE ART AND SCIENCE OF HOLDUP MEASUREMENTS

The measurement of nuclear material held up in processing plants is both an art and a science. It is subject to the constraints of politics, economics, and health and safety requirements, as well as to the laws of physics. For the practitioner, the measurement process is often long and tedious and is performed under difficult circumstances, as suggested by Figure 20.5. The work combines the features of a detective investigation and a treasure hunt, as aptly described by Zucker and Degen (Ref. 18). In fact, the cost of a thorough holdup measurement campaign is in the range of \$10 per gram of detected material—comparable to the price of gold.

Nuclear material held up in pipes, ducts, gloveboxes, heavy equipment, floors, walls, and so forth, is usually distributed in a diffuse and irregular manner. It is difficult for the assayist to define the measurement geometry, identify the form of the material, and measure it without interference from adjacent sources of radiation. For these reasons holdup measurement is an art that requires experience, imagination, a sense of proportion, and luck.

Holdup measurement also requires a scientific knowledge of radiation sources and detectors, calibration procedures, geometry, and error analysis. These topics are discussed in the remainder of this chapter.

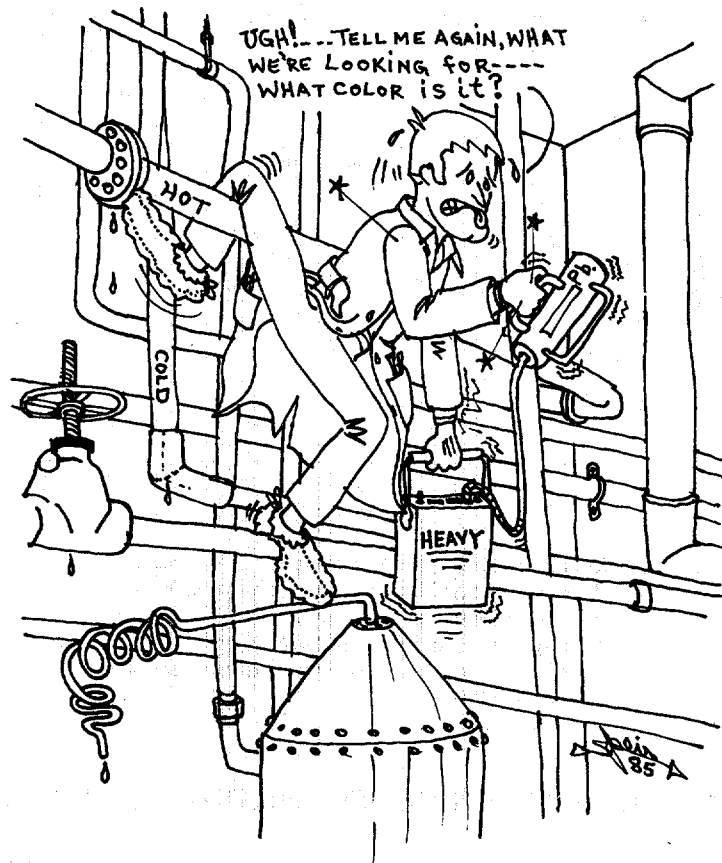


Fig. 20.5 Holdup measurements are both an art and a science and, as this cartoon illustrates, are usually conducted under difficult circumstances.

20.6.1 Useful Radiation Signatures

Table 20-4 lists gamma-ray and neutron radiation sources that are penetrating enough to measure holdup of uranium and plutonium. The tabulated neutron energies are approximate averages for the actual spontaneous fission, induced fission, or (α ,n) reaction neutron spectra. The neutron intensities are approximate values (calculated from Tables 11-1 and 11-3 in Chapter 11) that indicate the relative ease or difficulty of assaying various isotopes or compounds.

Most holdup measurements of uranium and plutonium are based on passive detection of the 186-keV ^{235}U gamma-ray peak and the 375- to 414-keV ^{239}Pu complex. Both portable sodium iodide detectors and thermoluminescent dosimeters (TLDs) are usually set up to respond to these gamma rays because they are the highest energy (and therefore the most penetrating) gamma rays available at useful intensities. These

Table 20-4. Useful radiation signatures for holdup measurements

Isotope	Technique	Signature	Intensity
^{235}U	passive gamma	186-keV gammas	43 000 $\gamma/\text{g-s}$
	active neutron	1-MeV neutrons	
UO_2	passive neutron	2-MeV neutrons	0.03 n/g-s ^a
UO_2F_2	passive neutron	1-MeV neutrons	2.0 n/g-s ^a
UF_6	passive neutron	1-MeV neutrons	5.8 n/g-s ^a
^{238}U	passive gamma	1001-keV gammas	100 $\gamma/\text{g-s}$
Pu	passive heat	infrared	
^{239}Pu	passive gamma	414-keV gammas	34 000 $\gamma/\text{g-s}$
		375-keV gammas	36 000 $\gamma/\text{g-s}$
		129-keV gammas	140 000 $\gamma/\text{g-s}$
^{240}Pu	passive neutron	2-MeV neutrons	1000 n/g-s
PuO_2	passive neutron	2-MeV neutrons	120 n/g-s ^b
PuF_6	passive neutron	1-MeV neutrons	7300 n/g-s ^b

^aHigh-enriched uranium with 1% ^{234}U .

^bLow-burnup plutonium with 0.03% ^{238}Pu , 6.5% ^{240}Pu , 92.5% ^{239}Pu .

intensities are sufficient to measure holdup with a sensitivity of 1 g. When uranium is mixed with thorium, measurement of the 186-keV ^{235}U peak may be difficult because of interferences from 200- to 300-keV radiation from thorium daughters (Ref. 19). When plutonium is measured with sodium iodide detectors, it is customary to set a window from 375 to 450 keV. This window will collect most 414-keV gamma rays and many 375-keV gamma rays but will exclude 332-keV gamma rays from ^{241}Pu or ^{241}Am (Ref. 13).

Large quantities of ^{238}U can be assayed with the low-intensity but very penetrating 1001-keV gamma rays from $^{234\text{m}}\text{Pa}$, a daughter of ^{238}U . After chemical separation of uranium, about a hundred days are required for the activity to come into equilibrium at the intensity given in Table 20-4.

Passive neutron counting may be helpful when it is necessary to measure holdup in pumps, valves, or other heavy equipment that is too dense to permit the escape of gamma rays. Neutrons penetrate metal and large holdup deposits better than gamma rays do, but they require more nuclear material to produce a strong signal. Neutron measurements are more difficult to interpret because neutrons do not have a unique energy, are difficult to collimate, are subject to multiplication and moderation effects, and can be increased in number by (α, n) reactions in chemical compounds. These effects cause neutron measurements to overestimate the amount of holdup, whereas self-attenuation effects cause gamma-ray measurements to underestimate the amount of holdup.

As indicated in Table 20-4, passive neutron counting of uranium in oxide or fluoride form is possible for reasonably large quantities. The neutron signal is due to (α, n) reactions in oxide or fluoride compounds. Active assay of ^{235}U is also possible but is very sensitive to nearby reflectors, moderators, and absorbers, and the response is

roughly proportional to the inverse fourth power of the uranium-to-instrument distance (Ref. 20). Passive neutron measurements of plutonium are quite practical, with spontaneous fission in ^{240}Pu being the primary neutron source unless large quantities of fluoride compounds are present.

The possible use of infrared scanning devices to locate plutonium holdup is being studied at several facilities. The primary source of infrared radiation is the heat generated in the alpha decay of ^{238}Pu . Infrared scanning of equipment to locate "hot spots" is probably feasible, but it is not known if quantitative measurements can be made.

20.6.2 Detectors and Readout Instrumentation

Because of the difficult circumstances under which holdup measurements must be carried out, the instrumentation should be portable, rugged, and easy to use. Simple push-button operation and low power consumption is also desirable in case battery-powered operation or operation inside plastic bags is necessary. All these requirements are satisfied by most of the equipment used today, which consists primarily of survey meters, portable multichannel analyzers, and collimated sodium iodide detectors.

Survey meters are geiger counters or small collimated or uncollimated sodium iodide detectors that respond to beta or gamma radiation. They are used for rapid surveys of large areas to locate holdup concentrations, and are usually equipped with audible rate meters. Quantitative holdup measurements are then performed with sodium iodide detectors (see Figure 20.6). The resolution and efficiency of these detectors, about 7 to 10%, is usually adequate for holdup measurements. Typical crystal sizes are 5-cm diam by 5-cm depth for plutonium measurements and 5-cm diam by 1-cm depth for uranium measurements. The detectors are collimated to limit their field of view. The collimators should consist of about 1.5 cm of lead for plutonium measurements and 0.5 cm of lead for uranium measurements in order to absorb at least 98% of the incident radiation (Refs. 13 and 21). An equivalent thickness of lead should cover the back of the detector to reduce background radiation. Also, the front face of the detector should be covered with 1.5 mm of lead (for plutonium measurements) and 0.8 mm of cadmium (for uranium and plutonium measurements, placed inside the lead) to reduce the count rate from x rays.

Commercially available electronics packages for sodium iodide detectors include the Eberline Stabilized Assay Meter (Chapter 4), the Brookhaven Stabilized Assay Meter (Ref. 18), and the Rocky Flats Assay Meter (Ref. 22), which uses Ludlum electronics. These packages are all portable, stabilized dual-channel analyzers. Portable multichannel analyzers are useful for identifying holdup of unknown composition and are now becoming available in packages small enough to serve as replacements for the stabilized assay meters. Commercial instruments include the Nuclear Data ND-6, the Canberra Series 10, and the Davidson Model 2056-4K.

The shielded neutron assay probe detector described in Chapter 15 can be used for neutron measurements of holdup (Ref. 23). The low front-to-back detection ratio, which ranges from 2 to 4, requires careful attention to background and the use of a collimator plug shield (Ref. 13). In general, 10 to 15 cm of polyethylene will reduce the intensity of

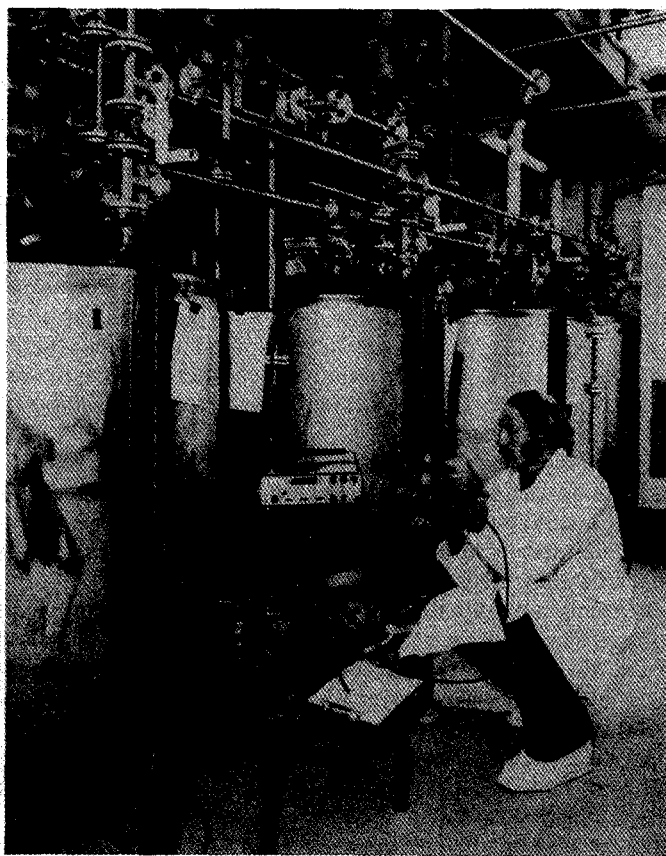


Fig. 20.6 An example of the use of a collimated sodium iodide detector and a portable electronics package to measure holdup in Raschig-ring filled tanks.

fission neutrons by a factor of 10. Large slab detectors can be employed for holdup measurements where portability is not a concern. Slab detectors have been used to estimate total room holdup (Ref. 24) and holdup in large calciners (Ref. 25).

Thermoluminescent dosimeters (TLDs) have been placed around the outside of gloveboxes (Ref. 26) and in the otherwise inaccessible interiors of calciners (Ref. 27). Lithium fluoride and calcium fluoride are common TLD crystals. Lithium fluoride has the advantage of greater availability, whereas calcium fluoride has a higher sensitivity and does not require a complex annealing cycle. For either material a graded shield is required to discriminate against low-energy x rays.

20.6.3 Holdup Measurement Procedures

Planning holdup measurements begins with several technical and nontechnical considerations. One important factor is whether or not measurements are to be periodic, routine, and can be started before hot operation of the facility commences. Where possible, measurements in advance of hot operation are very helpful in defining holdup collection zones, measuring backgrounds, measuring attenuation through equipment walls, and calibrating for difficult geometries. Brackets, fixtures, and special shields can be manufactured and installed for later use. Another important consideration is whether the holdup measurements are to be absolute or relative. Absolute measurements are more desirable because they yield values for the total grams held up in the facility. However, a change in holdup is easier to measure than the actual holdup because systematic errors tend to cancel. Periodic relative measurements may be sufficient for monitoring routine cleanout operations or for ensuring that holdup is not affecting the plant's monthly inventory balance.

The holdup measurement campaign itself consists of the following steps:

- (1) The measurement team studies the plant process and consults with plant operators to identify areas of potentially high holdup.
 - (2) A quick radiation survey with collimated or uncollimated survey meters indicates those areas where most material is held up.
 - (3) Most of the remaining measurement time is allocated to those areas with the majority of material. Other areas are measured more lightly or estimated by extrapolation. Note however that large areas with low holdup per unit area may contain large amounts of material. Conversely, localized hot spots may contain relatively small absolute quantities.
 - (4) The holdup detectors are collimated and calibrated using known standards. Small check sources are used to monitor instrument stability. Each detector is calibrated for point, line, and area collection zones.
 - (5) Quantitative measurement of holdup in the facility begins at this point. Each item of equipment to be measured is characterized as a point, line, or area holdup collection zone. The field of view of the detector is limited so that each collection zone can be resolved from its neighbors and from the background. The measurement team records the date, time, counting interval, collection zone identification, assumed collection geometry, source-to-detector distance, type and thickness of intervening material, and count rate. Each holdup measurement should be long enough to yield several hundred or several thousand counts. Then a background measurement is made by using a collimator plug or a movable shield or by moving the detector sideways so it misses the zone but views the same background.
 - (6) To obtain an estimate of uncertainty, the collection zone should be measured from a different direction, from a different distance, by assuming a different geometry (for example, point vs area), or by using a different measurement technique. In this matter the judgment and experience of the team members are paramount. They must guess at the distribution of material and choose measurement distances and calibration geometries accordingly in order to average holdup fluctuations and use their time to best advantage.
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(7) Although attenuation corrections, gram values of holdup, and error estimation can be calculated later, the team members should do some rough calculations on the spot. This is very important to ensure that they are spending their time where it is most needed and are not making large measurement errors.

20.6.4 Point, Line, and Area Calibrations

During a measurement campaign each holdup collection zone is characterized as a point, line, or area source so that the observed count rate can be easily converted to grams of nuclear material. For example; a pump, filter, or valve may be considered a point source if the holdup is distributed over distances that are small compared to the source-to-detector distance and if the holdup is entirely within the detector field of view; a long pipe or duct may be considered a line source; a wall, floor, or broad rectangular duct that extends well beyond the detector field of view may be considered an area source. Sometimes the choice of point, line, or area calibration is not obvious and is a matter of judgment or experience. Sometimes the measurement team may try two or even all three possibilities and compare the final results for the holdup. Or the team may measure the count rate as a function of distance from the collection zone to help establish the proper calibration choice. For a point source, the measured response falls off as the inverse square of the distance ($1/r^2$). For a line source, the response falls off as the inverse of the distance ($1/r$). For a uniform area source, the response is independent of distance. This last case is not as obvious as it seems; it is predicated on the finite viewing angle of the collimated detector, which views an area that increases as r^2 , thereby canceling the $1/r^2$ falloff in response with distance.

Point, line, and area calibrations can of course be obtained from point sources, line sources, and area sheet standards, as described in Section 20.6.5. However, it is also possible, and usually easier, to obtain all three calibrations from a single point source. The procedure for doing this with a gamma-ray detector is described below.

(1) Collimate the detector by recessing it in its lead shield by one or two crystal diameters to obtain a viewing half-angle $\theta/2$ of 15 to 30 degrees (see Figure 20.7). This collimation must now remain fixed because the line and area calibration constants are strongly dependent on the field of view.

(2) Place the point calibration source at a fixed distance r_0 (typically 1 to 2 m). Determine the count rate C_0 . Now move the source sideways in fixed steps of width s (typically 10 to 20 cm), as illustrated in Figure 20.7. Determine the count rate C_i at each step i , with each count rate corrected for background. The result is a response curve similar to the example in Figure 20.8.

(3) The curve of detector response as a function of sideways displacement falls off because of the finite viewing angle of the collimated detector. If the collimation were perfect, with viewing half-angle $\theta/2$, the detector could view a length $L \approx r_0\theta$ of a line source or an area $A \approx \pi r_0^2\theta^2/4$ of an area source. Actually, the equivalent length L of a uniform line source that gives the same count rate as the integrated response curve is

$$L = 2s \sum C_i/C_0 \quad (20-1)$$

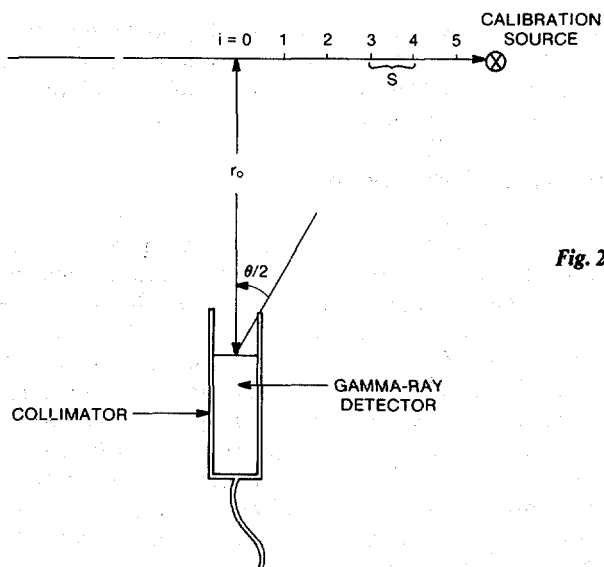
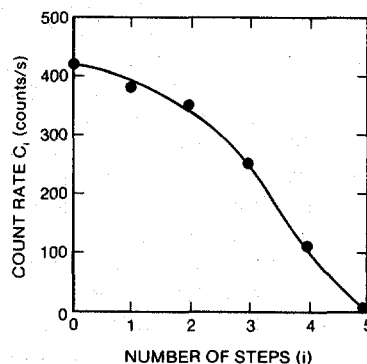


Fig. 20.7 The geometry required for calculating line and area calibrations. At a fixed distance r_0 the point calibration source is moved sideways in fixed steps s .

Fig. 20.8 Collimated gamma-ray detector response as a function of sideways displacement in number of steps. The response falls off because of the finite viewing angle of the collimated detector. The equivalent length L and equivalent area A viewed by the detector can be calculated from this curve.



(4) The equivalent area A of a uniform area source that gives the same count rate as the integrated response curve is

$$A = \sum a_i C_i / C_0 \quad (20-2)$$

To obtain this equation we have imagined that at each sideways position i of the point source, the measured response is representative of that which would be obtained over an annular ring of inner radius $(i - 1/2)s$ and outer radius $(i + 1/2)s$. The area a_i of each annulus is $2i\pi s^2$, except that $a_0 = \pi s^2/4$.

(5) If the point source standard contains m_0 grams of nuclear material, the point calibration for holdup is

$$m(\text{holdup}) = m_0 \frac{C}{C_0} \frac{r^2}{r_0^2} \text{ (grams)} \quad (20-3)$$

where C is the observed count rate corrected for background, r is the detector-to-holdup distance, and m is the mass of holdup in grams.

(6) The line calibration is

$$m(\text{holdup}/m) = \frac{m_0}{L} \frac{C}{C_0} \frac{r}{r_0} \text{ (g/m)} \quad (20-4)$$

C_0 must be measured at the distance r_0 used to determine L .

(7) The area calibration is

$$m(\text{holdup}/m^2) = \frac{m_0}{A} \frac{C}{C_0} \text{ (g/m}^2\text{)} \quad (20-5)$$

C_0 must be measured at the distance r_0 used to determine A .

(8) Note that the preceding equations assume that the same standard, of mass m_0 , is used to determine L , A , and C_0 . This is convenient in practice but not essential. One standard could be used for the measurements required to calculate L and A with Equations 20-1 and 20-2, and another could be used to provide m_0 and C_0 for Equations 20-3 through 20-5.

20.6.5 Calibration Standards and Check Sources

In principle, the geometry of a calibration standard should be the same as the geometry of the unknown being measured. For holdup measurements this is usually not possible. Therefore, in practice, point, line, and area standards are used to approximate equipment geometries. Also, as described in the preceding section, a single small calibration standard can be used to obtain point, line, and area calibrations. For gamma-ray measurements, 1 to 5 g of ^{235}U or ^{239}Pu is sufficient. Even for these small gamma-ray standards, self-absorption is significant and must be corrected for (see, for example, Table 20-5). For neutron measurements, 10 to 20 g of plutonium (6 to 20% ^{240}Pu) gives an adequate count rate. In the neutron standard, self-absorption and self-multiplication are negligible, but it is important to establish by calculation or measurement that the neutron production rate in the standard is representative of actual plant material.

To ensure the stability and reliability of portable radiation detectors in a plant environment, it is necessary to carry along small check sources. The performance of the detectors should be checked against these sources every 1 to 4 h. The point calibration standards described in the preceding paragraph (or even somewhat smaller sources) are suitable for this application.

Some holdup measurement teams have fabricated sheet standards to supplement their point calibration standards. One common technique involves sprinkling oxide powder on transparent plastic sheets coated with adhesive (Ref. 18). Uranium oxide has also been mixed with silicon rubber and deposited on sheets (Ref. 19). The sheet standards can be used for area calibrations or rolled up in pipes and ducts for line calibrations. The sheet standards may be difficult to fabricate or use, however, because the oxide may be deposited nonuniformly and may become stiff, causing it to crack or flake.

In any facility there may be special material holdup geometries that cannot be approximated by point, line, or area sources. Sometimes it is possible to mock up these geometries with combinations of sheet standards and point standards. Another alternative is to put known standards inside the actual process equipment, although this can usually be done only before the equipment is placed into operation.

20.6.6 Self-Absorption and Attenuation Corrections

A chronic problem in passive gamma-ray holdup measurements is the tendency to underestimate holdup because of self-absorption in the material itself or attenuation in intervening materials. Self-absorption of the gamma rays in uranium or plutonium can be very severe, as indicated by the two examples in Table 20-5. Although the assayist cannot correct for self-absorption because the density and distribution of the material are unknown, he may be able to make some allowance for self-absorption in estimating errors.

Table 20-5. Estimated self-absorption and attenuation corrections for common materials encountered in holdup measurements

Intervening Material	Correction for 186-keV Gamma Rays	Correction for 414-keV Gamma Rays
1-g-cube UO_2	2.95	
10-g-cube UO_2	5.97	
1-g-cube PuO_2		1.28
10-g-cube PuO_2		1.66
Rubber glove	1.05	1.04
0.25-in. Plexiglas	1.11	1.08
1.0-in. water	1.42	1.30
0.25-in. aluminum	1.24	1.18
8-in. \times 8-in. HEPA filter	1.43	1.31
0.063-in. steel	1.20	1.12
0.125-in. steel	1.44	1.25
0.250-in. steel	2.08	1.55
0.063-in. lead	6.83	1.44

Attenuation of the gamma rays by intervening pipe walls, gloveboxes, or other materials can be determined by calculation or by transmission measurements. A transmission measurement is illustrated in Figure 20.9, where a source (typically ^{137}Cs) is positioned behind a duct. The measurement procedure, calculation of transmission, and conversion to a self-attenuation correction factor is the same as that given in Chapter 6. The procedure is rarely used during a holdup measurement campaign because it is time-consuming, physically awkward, and requires different electronic settings to measure the 662-keV cesium peak. Instead, it is usually sufficient to calculate the attenuation by estimating the thickness and composition of the intervening materials.

Table 20-5 provides examples of gamma-ray transmission through common materials and the associated attenuation correction. Although in practice the attenuation correction is only an estimate, it is very important to make this estimate for *every* holdup measurement until it is known by experience where the correction can be neglected. Otherwise the holdup measurement is merely a lower limit on the amount of material actually present.

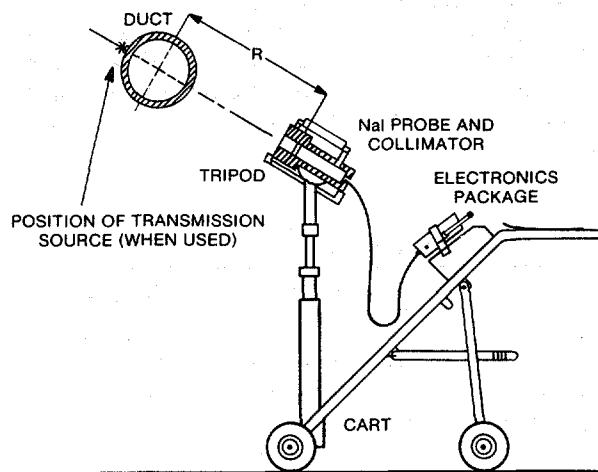


Fig. 20.9 Holdup measurement of a duct showing placement of transmission source for attenuation correction measurement.

20.6.7 Error Estimation

Both the art and science of holdup measurements are involved in the process of estimating measurement errors. These errors are large and numerous; their causes are summarized below in a somewhat subjective ordering of decreasing importance:

- (1) Unknown material distribution, which affects the source-to-detector distance and the validity of the chosen point, line, or area calibration.
- (2) Self-absorption in the material or its matrix.
- (3) Gamma-ray attenuation by intervening materials.

- (4) Background interference from distant line-of-sight objects or from adjacent, unresolved material.
- (5) Detector instability or improper calibration.
- (6) Unrepresentative standards.
- (7) Statistical imprecision.
- (8) Uncertainty in material isotopic composition.

Statistical imprecision is the only source of error that can be treated in a rigorous fashion; it is usually negligible compared with other errors.

The most important technique for error estimation available to the measurement team is that of measuring each collection zone in several different ways. After each measurement is properly corrected for distance, background, attenuation, and so forth, and a gram value for holdup is obtained, the different values should be averaged together. The measurement standard deviation can be estimated or calculated from the range of values.

To estimate the accuracy of a series of holdup measurements, the holdup must be measured both before and after a cleanout campaign. If the actual amount of material removed can be determined by sampling and chemical analysis or by other nondestructive assay techniques, then the measurement accuracy can be calculated. Previous calibrations can be updated, and error estimates can be reassessed.

Table 20-6 gives a brief summary of published comparisons of holdup measurements and cleanout campaigns in existing facilities; the overall accuracy of holdup measurements can be estimated from the data. In general, the accuracy of holdup measurements is $\pm 50\%$, although better results can be obtained for favorable geometries or carefully controlled measurement campaigns.

Table 20-6. Typical accuracy of holdup measurements

Reference	Material	Location	Measurement Technique	Accuracy (\pm) (%)
19	HEU oxide	ducts	passive gamma	10-20
		filters	passive gamma	50-100
23	PuO ₂	gloveboxes	passive gamma	10
			passive neutron	15
24	PuO ₂	gloveboxes	passive gamma	50
		total rooms	passive neutron	50
26	PuO ₂	gloveboxes	TLDs	20
14	HEU oxide	ducts	passive gamma	20
		precipitator	passive gamma	15
		calciners	passive gamma	20
		pipes	passive gamma	10
		pumps	passive gamma	25
28	UF ₆	enrichment cascade	passive gamma	50

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1. The first part of the document is a letter from the President of the United States to the Congress, dated January 3, 1862. It is a very important document, as it contains the President's message to the Congress, and is a very important document, as it contains the President's message to the Congress.

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